CLAIMS

A surface-protected plastic composite material comprising a transparent plastic, a coating layer (I) as
 a first layer stacked on the transparent plastic substrate and a thermally cured coating layer (II) as a second layer stacked on the first layer, the coating layer (I) being formed of a resin composition containing at least 50 % by weight, based on the resin content
 thereof, of an acrylic resin which is an acrylic resin containing at least 50 mol% of recurring unit of the following formula (I-a),

$$\begin{array}{c}
CH_3 \\
-[CH_2 - C] - \\
COOR^1
\end{array}$$
(I-a)

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wherein R^1 is an alkyl group having 1 to 4 carbon atoms,

the thermally cured coating layer (II) being made of an organosiloxane resin formed from the following components a, b and c,

- (A) colloidal silica (component a),
- (B) a hydrolysis condensate (component b) of a trialkoxysilane of the following formula (II-1),

$$R^2Si(OR^3)_3$$
 (II-1),

wherein R² is an alkyl group having 1 to 4 carbon atoms, a vinyl group or an alkyl group which has 1 to 3 carbon atoms and is substituted with at least one group selected from the group consisting of methacryloxy, amino, glycidoxy and 3,4-epoxycyclohexyl and R³ is an

30 alkyl group having 1 to 4 carbon atoms, and (C) a hydrolysis condensate (component c) of a tetraalkoxysilane of the following formula (II-2),

$$Si(OR^4)_4$$
 (II-2)

wherein R^4 is an alkyl group having 1 to 4 carbon

atoms, the organosiloxane resin containing 5 to 45 % by weight of the component a, 50 to 80 % by weight, as $R^2SiO_{3/2}$, of the component b and 2 to 30 % by weight, as SiO_2 , of the component c.

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- 2. The transparent plastic composite material of claim 1, wherein the acrylic resin of the coating layer (I) is an acrylic copolymer resin having recurring units of the said formula (I-a) and the following formula (I-b)
- 10 and/or the following formula (I-c),

in which X is a hydrogen atom or methyl, R⁵ is an alkylene group having 2 to 5 carbon atoms, R⁶ is an alkyl group having 1 to 4 carbon atoms and n is an integer of 0 or 1,

$$-[CH_2 - C] - CC$$
 (I-c)

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in which Y is a hydrogen atom or methyl and R^9 is an alkylene group having 2 to 5 carbon atoms,

and further wherein the acrylic copolymer resin has a (I-a):[(I-b) + (I-c)] molar ratio of the units in the range of from 99.5:0.5 to 50:50.

3. The transparent plastic composite material of claim 2, wherein the acrylic copolymer resin is an acrylic copolymer resin containing 0.3 to 40 mol%, based on the total units of the above formulae (I-a) and [(I-b) + (I-c)], of units of the following formula (I-d),





wherein Z is a hydrogen atom or methyl, R10 is an alkylene group having 2 to 5 carbon atoms and W is an ultraviolet absorbent residue.

The transparent plastic composite material of any 5 one of claims 1 to 3, wherein the coating layer (I) is formed of a mixture of the acrylic resin with a hydrolysis condensate of a compound of the following formula (I-e),

 R^7_r -Si(OR⁸)_{4-r} 10 (I-e)

> wherein R⁷ is an alkyl group having 1 to 4 carbon atoms, a vinyl group or an alkyl group which is substituted with one or more groups selected from the class consisting of methacryloxy, amino, glycidoxy and 3,4-epoxycyclohexyl and has 1 to 3 carbon atoms, R^8 is an alkyl group having 1 to 4 carbon atoms, and r is an integer of 0 to 2, the mixture having an acrylic resin:hydrolysis condensate (as R_r^7 -SiO_{4-r/2}) amount ratio by weight in the range of from 99:1 to 60:40,

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- The transparent plastic composite material of claim 4, wherein the coating layer (I) further contains a melamine resin in the range of from 1 to 20 parts by weight per 100 parts by weight of the total of the 25 acrylic resin and the hydrolysis condensate of the compound of the formula (I-e).
 - 6. The transparent plastic composite material of any one of claims 1 to 4, wherein the coating layer (I) further contains 0.7 to 100 parts by weight, per 100 parts by weight of the acrylic resin, of an ultraviolet absorbent.
 - 7. The transparent plastic composite material of claim

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- 6, wherein the ultraviolet absorbent has a solubility of 1.5 g/L or less in a solvent mixture containing 100 parts by weight of isopropanol, 30 parts by weight of methanol and 15 parts by weight of water when measured at 25°C.
- The transparent plastic composite material of claim 1, wherein the coating layer (II) is a thermally cured layer of a resin composition containing 15 to 35 % by weight of the component a, 55 to 75 % by weight, as R²SiO_{3/2}, of the component b and 3 to 20 % by weight, as SiO₂, of the component c.
- 9. The transparent plastic composite material of claim
 15 1, wherein:
 - (1) the coating layer (I) is formed of a resin composition containing an acrylic copolymer resin which is formed of recurring units of the following formulae (I-a) and (I-b),

$$\begin{array}{c}
CH_3 \\
--[CH_2 - C] - \\
COOR^1
\end{array}$$
(I-a)

wherein R^1 is as defined in the foregoing formula (I-a),

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$$X$$
—[CH₂ — C]— (I-b)
COO — R⁵ — Si(CH₃)_n(OR⁶)_{3-n}

wherein X, R^5 , R^6 and n are as defined in the foregoing formula (I-b),

and which has a (I-a):(I-b) unit ratio in the range of from 97:3 to 70:30 and 3 to 60 parts by weight, per 100 parts by weight of the acrylic copolymer resin, of an ultraviolet absorbent, the ultraviolet absorbent having a solubility of 1.5 g/L or less in a solvent mixture

containing 100 parts by weight of isopropanol, 30 parts by weight of methanol and 15 parts by weight of water when measured at 25°C, and

- (2) the coating layer (II) is a thermally cured layer of a resin composition containing 15 to 35 % by weight of the component a, 55 to 75 % by weight, as $R^2SiO_{3/2}$, of the component b and 3 to 20 % by weight, as SiO_2 , of the component c.
- 10 10. The transparent plastic composite material of claim 1, wherein:
 - (1) the coating layer (I) is formed of a resin composition containing an acrylic copolymer resin which is formed of recurring units of the following formulae (I-a), (I-b) and (I-d),

$$\begin{array}{c}
CH_3 \\
--[CH_2 - C] - \\
COOR^1
\end{array}$$
(I-a)

wherein R^1 is as defined in the foregoing formula 20 (I-a),

$$\begin{array}{c}
X \\
--[CH_2 - C] - \\
COO - R^5 - Si(CH_3)_n(OR^6)_{3-n}
\end{array}$$

wherein X, R^5 , R^6 and n are as defined in the 25 foregoing formula (I-b),

$$--[CH2 - C - C] - (I-d)$$

$$COO - R10 - W$$

wherein Z, R^{10} and W are as defined in the foregoing 30 formula (I-d),

and which has a (I-a):(I-b):(I-d) unit ratio in the range of from 96.7-60:3-30:0.3-15 and 3 to 60 parts by weight, per 100 parts by weight of the acrylic copolymer resin, of an ultraviolet absorbent, the ultraviolet

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absorbent having a solubility of 1.5 g/L or less in a solvent mixture containing 100 parts by weight of isopropanol, 30 parts by weight of methanol and 15 parts by weight of water when measured at 25°C, and

- (2) the coating layer (II) is a thermally cured layer of a resin composition containing 15 to 35 % by weight of the component a, 55 to 75 % by weight, as $R^2SiO_{3/2}$, of the component b and 3 to 20 % by weight, as SiO_2 , of the component c.
- 11. The transparent plastic composite material of claim 1, wherein:
- (1) the coating layer (I) is formed of a resin composition containing 100 parts by weight of an acrylic copolymer resin which is formed of recurring units of the following formulae (I-a) and (I-c),

$$\begin{array}{c}
CH_3 \\
--[CH_2 - C] - \\
COOR^1
\end{array}$$
(I-a)

wherein R^1 is as defined in the foregoing formula (I-a),

$$-[CH_2 - C] - COO - R^9 - OH$$
 (I-c)

wherein Y and R^9 are as defined in the foregoing formula (I-c),

and which has a (I-a):(I-c) unit ratio in the range of from 95:5 to 60:40, a mixture (as $R^7_rSiO_{4-r/2}$) of hydrolysis condensates of a compound of the formula (I-e),

$$R^{7}_{r}$$
-Si(OR⁸)_{4-r} (I-e)

wherein R^7 , R^8 and r are as defined in the foregoing formula (I-e), the mixture having such an amount that the ratio of the acrylic copolymer resin:the hydrolysis

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condensates comes to be 99:1 to 60:40 by weight, a melamine resin in such an amount that the amount thereof per 100 parts by weight of the total of the acrylic copolymer resin and the hydrolysis condensates comes to be 3 to 15 parts by weight, and 3 to 60 parts by weight, per 100 parts by weight of the acrylic copolymer resin, of an ultraviolet absorbent, the ultraviolet absorbent having a solubility of 1.5 g/L or less in a solvent mixture containing 100 parts by weight of isopropanol, 30 parts by weight of methanol and 15 parts by weight of water when measured at 25°C, and

- (2) the coating layer (II) is a thermally cured layer of a resin composition containing 15 to 35 % by weight of the component a, 55 to 75 % by weight, as $R^2SiO_{3/2}$, of the component b and 3 to 20 % by weight, as SiO_2 , of the component c.
- 12. The transparent plastic composite material of claim 1, wherein:
- (1) the coating layer (I) is formed of a resin composition containing 100 parts by weight of an acrylic copolymer resin which is formed of recurring units of the following formulae (I-a), (I-c) and (I-d),

$$\begin{array}{ccc}
 & CH_3 \\
 & CH_2 & C \\
 & COOR^1
\end{array}$$
(I-a)

wherein R^1 is as defined in the foregoing formula (I-a),

$$-[CH_2 - C] - C$$
 $-[CH_2 - C] - C$
 $-[CH_2 - C] - C$
 $-[CH_2 - C] - C$
 $-[CH_2 - C] - C$

wherein Y and R^9 are as defined in the foregoing formula (I-c),

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wherein Z, R^{10} and W are as defined in the foregoing 5 formula (I-d),

and which has a (I-a):(I-c):(I-d) unit ratio in the range of from 94.7-50:5-40:0.3-15, a mixture (as $R_{r}^{7}SiO_{4-}$ r/2) of hydrolysis condensates of a compound of the formula (I-e),

$$R^{7}_{r}-Si(OR^{8})_{4-r} \qquad (I-e)$$

wherein R⁷, R⁸ and r are as defined in the foregoing formula (I-e), the mixture having such an amount that the ratio of the acrylic copolymer resin: the hydrolysis condensates comes to be 99:1 to 60:40 by weight, a melamine resin in such an amount that the amount thereof per 100 parts by weight of the total of the acrylic copolymer resin and the hydrolysis condensates comes to be 3 to 15 parts by weight, and 3 to 60 parts by weight, per 100 parts by weight of the acrylic copolymer resin, of an ultraviolet absorbent, the ultraviolet absorbent having a solubility of 1.5 g/L or less in a solvent mixture containing 100 parts by weight of isopropanol, 30 parts by weight of methanol and 15 parts by weight of water when measured at 25°C, and

(2) the coating layer (II) is a thermally cured 25 layer of a resin composition containing 15 to 35 % by weight of the component a, 55 to 75 % by weight, as $R^2SiO_{3/2}$, of the component b and 3 to 20 % by weight, as SiO_2 , of the component c.

The transparent plastic composite material of claim 1, wherein the transparent plastic substrate is made of an aromatic polycarbonate resin.

- 14. The transparent plastic composite material of claim 1, which has a thickness in the range of from 0.001 to 10 mm.
- 5 15. The transparent plastic composite material of claim 1, wherein, when subjected to a 1,000-rotation Taber abrasion test (ASTM D1044) using a CS-10F abrasion wheal supplied by Calibrase under a load of 500 g, the transparent plastic composite material shows a change of 10 2 % or less between its haze values before and after the test.
- 16. The transparent plastic composite material of claim 1, which substantially does not show any occurrence of cracking on the surface thereof after maintained in boiling water for 3 hours.
 - 17. An organic window glass made of the transparent plastic composite material recited in claim 1.
 - 18. An organic window glass for a car or an aircraft, which is made of the transparent plastic composite material recited in claim 1.

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